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For and on behalf of RWS Group Ltd

The 20th day of August 2004

A sealable biaxially oriented polyester film with one matt side

The invention relates to a sealable biaxially oriented polyester film with one matt side and composed of at least one base layer (B) and, applied to the two sides of this base layer (B), outer layers (A) and (C). The invention further relates to a process for the production of the film, and to use of the film.

Sealable, biaxially oriented polyester films are known from the prior art.

GB-A 1 465 973 describes a coextruded polyester film having two layers, one layer of which consists of copolyesters containing isophthalic acid and terephthalic acid, and the other layer of which consists of polyethylene terephthalate. The lack of pigmentation means that the film cannot be produced by a reliable process (cannot be wound up) and that the possibilities for further processing of the film are limited.

EP-A 0 035 835 describes a coextruded, sealable polyester film where, in the sealable layer, particles whose average size exceeds the sealable layer thickness are present in order to improve winding and processing performance. The particulate additives form surface protrusions which prevent undesired blocking and sticking of the film to rolls or guides. No further details are given concerning the incorporation of antiblocking agents in relation to the other, nonsealable layer of the film. It is uncertain whether

this layer comprises antiblocking agents. The patent does not give any indication of the sealing temperature range of the film. The seal seam strength is measured at 140 °C and is in the range from 63 to 120 N/m (from 0.97 N/15 mm to 1.8 N/15 mm of film width).

EP-A 0 432 886 describes a coextruded multilayer polyester film which has a first surface on which has been arranged a sealable layer, and has a second surface on which has been arranged an acrylate layer. The sealable outer layer here may also be composed of isophthalic-acid-containing and terephthalic-acid-containing copolyesters. The coating on the reverse side gives the film improved processing performance. The patent gives no indication of the sealing temperature range of the film. The seal seam strength is measured at 140 °C. For a sealable layer thickness of 11 µm the seal seam strength given is 761.5 N/m (11.4 N/15 mm of film width). A disadvantage of the reverse-side acrylate coating is that this side is now not sealable with respect to the sealable outer layer, and the film therefore has only very restricted use.

EP-A 0 515 096 describes a coextruded, multilayer, sealable polyester film which comprises a further additive in the sealable layer. The additive may comprise inorganic particles, for example, and is preferably distributed in an aqueous layer onto the film during its production. Using this method, the film is claimed to retain its good sealing properties and to be easy to process. The reverse side of the film comprises only very few particles, most of which pass into this layer via the recycled material. This patent again gives no indication of the sealing temperature

range of the film. The seal seam strength is measured at 140 °C and is above 200 N/m (3 N/15 mm of film width). For a sealable layer of 3 mm thickness the seal seam strength given is 275 N/m (4.1 N/15 mm of film width).

WO 98/06575 describes a coextruded, multilayer polyester film which comprises a sealable outer layer and a nonsealable base layer. The base layer here may have been built up from one or more layers, and the inner layer of these layers is in contact with the sealable layer. The other (outward-facing) layer then forms the second nonsealable outer layer. Here, too, the sealable outer layer may be composed of isophthalic-acid-containing and terephthalic-acid-containing copolyesters, but these comprise no antiblocking particles. The film also comprises at least one UV absorber, which is added to the base layer in a ratio of from 0.1 to 10 % by weight. The base layer of this film has conventional antiblocking agents. Depending on the embodiment, the film may also be matt. The film features good sealability, but does not have the desired processing performance, and has shortcomings in optical properties (gloss and haze).

The prior art also discloses matt, milky, biaxially oriented polyester films.

DE-A 23 53 347 describes a process for the production of a single- or multilayer, milky polyester film, which comprises preparing a mixture of particles of a linear polyester with from 3 to 27 % by weight of a homopolymer or copolymer of ethylene or propylene, extruding the mixture in the form of a film, quenching

the film, and biaxially orienting it via orientation in directions running perpendicularly to one another, and heat-setting the film. A disadvantage of this process is that it is impossible to reuse regrind produced during the production of the film (in essence a mixture of polyester and ethylene copolymer or propylene copolymer), because otherwise the film becomes yellow. This makes the process uneconomic, and there was no market for the discolored film produced with regrind. On increasing the concentration of the copolymer in the polyester, the film generally loses its milky character and becomes white, with high opacity.

It was an object of the present invention to provide a sealable, biaxially oriented polyester film with one matt side and without the disadvantages of the films mentioned of the prior art, and in particular featuring excellent optical properties (high single-side mattness together with good transparency), very good sealability, and improved processability. In addition, the film is intended to be capable of processing in a reliable continuous operation, even on high-speed processing machinery. A further intention is to ensure that an amount of up to 60 % by weight, based on the total weight of the film, of chopped material produced in the factory during the production of the film can be reintroduced as regrind into the production process, without any significant resultant adverse effect on the physical and optical properties of the film.

The invention achieves the object by providing a sealable, biaxially oriented and coextruded polyester film with one matt side and with at least one base layer (B), with a sealable outer layer (A), and with a

matt outer layer (C), wherein

a) the sealable outer layer (A) has a minimum sealing temperature at most 120 °C, a seal seam strength of at least 1.0 N/15 mm of film width, an R_a value of at most 100 nm, and a value measured for surface gas flow in the range from 20 to 4000 s;
b) the matt outer layer (C) has a gloss of at most 100, an R_a value of at least 150 nm, and a value measured for surface gas flow in the range from 0 to 50 s, and
c) the haze is smaller than 50 %.

The subclaims give preferred embodiments of the invention, and these are described in more detail below.

According to the invention, the film has at least three layers, the layers then encompassed being the base layer (B), the sealable outer layer (A), and the nonsealable outer layer (C).

The base layer (B) of the film preferably comprises at least 90 % by weight of thermoplastic polyester. Polyesters suitable for this purpose are those made from ethylene glycol and terephthalic acid (polyethylene terephthalate, PET), from ethylene glycol and naphthalene-2,6-dicarboxylic acid (polyethylene 2,6-naphthalate, PEN), from 1,4-bishydroxymethylcyclohexane and terephthalic acid (poly(1,4-cyclohexanedimethylene terephthalate), PCDT), or else made from ethylene glycol, naphthalene-2,6-dicarboxylic acid and biphenyl-4,4'-dicarboxylic acid (polyethylene 2,6-naphthalate bibenzoate, PENBB). Particular preference is given to polyesters of which at least

90 mol%, preferably at least 95 mol%, is composed of ethylene glycol units and terephthalic acid units, or of ethylene glycol units and naphthalene-2,6-dicarboxylic acid units. The remaining monomer units derive from other aliphatic, cycloaliphatic or aromatic diols and, respectively, dicarboxylic acids, as may also occur in the layer (A), or in the layer (C).

Other examples of suitable aliphatic diols are diethylene glycol, triethylene glycol, aliphatic glycols of the general formula $HO-(CH_2)_n-OH$, where n is an integer from 3 to 6 (in particular 1,3-propanediol, 1,4-butanediol, 1,5-pentanediol and 1,6-hexanediol) and branched aliphatic glycols having up to 6 carbon atoms. Among the cycloaliphatic diols, mention should be made of cyclohexanediols (in particular 1,4-cyclohexanediol). Examples of other suitable aromatic diols have the formula $HO-C_6H_4-X-C_6H_4-OH$, where X is $-CH_2-$, $-C(CH_3)_2-$, $-C(CF_3)_2-$, $-O-$, $-S-$ or $-SO_2-$. In addition, bisphenols of the formula $HO-C_6H_4-C_6H_4-OH$ are also very suitable.

Other aromatic dicarboxylic acids are preferably benzenedicarboxylic acids, naphthalenedicarboxylic acids, such as naphthalene-1,4- or -1,6-dicarboxylic acid, biphenyl-x,x'-dicarboxylic acids, in particular biphenyl-4,4'-dicarboxylic acid, diphenylacetylene-x,x'-dicarboxylic acids, in particular diphenyl-acetylene-4,4'-dicarboxylic acid, or stilbene-x,x'-dicarboxylic acids. Among the cycloaliphatic dicarboxylic acids mention should be made of cyclohexanedicarboxylic acids, in particular cyclohexane-1,4-dicarboxylic acid. Among the aliphatic dicarboxylic acids, the (C_3-C_{19}) alkanediacids are

particularly suitable, and the alkane moiety here may be straight-chain or branched.

One way of preparing the polyesters is the transesterification process. Here, the starting materials are dicarboxylic esters and diols, which are reacted using the customary transesterification catalysts, such as the salts of zinc, of calcium, of lithium, of magnesium or of manganese. The intermediates are then polycondensed in the presence of well-known polycondensation catalysts, such as antimony trioxide or titanium salts. Another equally good preparation method is the direct esterification process in the presence of polycondensation catalysts. This starts directly from the dicarboxylic acids and the diols.

The sealable outer layer (A) applied by coextrusion to the base layer (B) has a structure based on polyester copolymers and in essence consists of copolyesters composed predominantly of isophthalic acid units and of terephthalic acid units, and of ethylene glycol units. The remaining monomer units derive from other aliphatic, cycloaliphatic or aromatic diols and, respectively, dicarboxylic acids, as may also occur in the base layer. Preferred copolyesters which provide the desired sealing properties are those which have been built up from ethylene terephthalate units and from ethylene isophthalate units and from ethylene glycol units. The proportion of ethylene terephthalate is from 40 to 95 mol%, and the corresponding proportion of ethylene isophthalate is from 60 to 5 mol%. Preference is given to copolyesters in which the proportion of ethylene terephthalate is from 50 to

90 mol% and the corresponding proportion of ethylene isophthalate is from 50 to 10 mol%, and particular preference is given to copolyesters in which the proportion of ethylene terephthalate is from 60 to 85 mol% and the corresponding proportion of ethylene isophthalate is from 40 to 15 mol%.

The desired sealing properties and the desired processing properties of the inventive film are obtained from the combination of the properties of the copolyester used for the sealable outer layer (A) and from the surface topographies, both of the sealable outer layer (A) and of the matt outer layer (C).

The desired minimum sealing temperature of 120 °C and the seal seam strength of at least 1.0 N/15 mm of film width is achieved if the copolymers described in more detail above are used for the sealable outer layer (A). To achieve the desired handling, and to achieve the good processing properties, the sealable outer layer (A) is further modified.

In one particularly advantageous embodiment, this is achieved with the aid of suitable antiblocking agents of a selected size, a certain amount of which is added to the sealable layer, and specifically in such a way as firstly to minimize the blocking of the film and secondly to prevent any significant impairment of the sealing properties. The antiblocking agents for the sealable outer layer (A) are in principle the same as those for the matt outer layer (C). However, when comparison is made with the matt outer layer (C), the amount of antiblocking agent is much smaller in the case of the sealable outer layer (A). To achieve the

abovementioned properties, the amount of antiblocking agent in the sealable outer layer (A) is from 0.01 to 1 % by weight, preferably from 0.015 to 0.08 % by weight, and in the particularly preferred embodiment is from 0.02 to 0.08 % by weight. The desired property combinations can be achieved particularly when the topography of the sealable outer layer (A) is described by the following parameters:

the roughness of the sealable outer layer, expressed via its R_a value, is to be smaller than or equal to 60 nm. Otherwise, the sealable properties are adversely affected for the purposes of the present invention. According to the invention, the value measured for surface gas flow is to be in the range from 20 to 4000 s. At values below 20 s, the sealing properties are adversely affected for the purposes of the present invention, while at values above 4000 s the handling of the film becomes markedly poorer.

In one advantageous embodiment, the coefficient of friction of the sealable outer layer (A) with respect to itself is at most 1.0, and in one particularly advantageous embodiment it is at most 0.8. In the particularly advantageous embodiment, the handling and the processing performance of the film are particularly good.

In order to achieve the desired mattness/the desired degree of mattness, the outer layer (C) comprises an effective amount of from 0.5 to 20 % by weight, based on the total weight of the outer layer (C), of particles (also termed additives or pigments). In the preferred embodiment of the film according to the

present invention, the particle concentration is from 1.0 to 18 % by weight, and in the particularly preferred embodiment it is from 1.5 to 16 % by weight.

Typical particles contributing to the degree of mattness of the film are inorganic and/or organic particles, such as calcium carbonate, amorphous silica, talc, magnesium carbonate, barium carbonate, calcium sulfate, barium sulfate, lithium phosphate, calcium phosphate, magnesium phosphate, aluminum oxide, lithium fluoride, the calcium, barium, zinc, or manganese salts of the dicarboxylic acids used, carbon black, titanium dioxide, kaolin, or crosslinked polymer particles, e.g. polystyrene or acrylate particles.

In addition, mixtures of two or more different particle systems or mixtures of particle systems of identical chemical composition but different particle size may also be selected. The particles may be added to the polymers of the individual layers of the film at the respective advantageous concentrations, e.g. in the form of a glycolic dispersion during the polycondensation process, or by way of masterbatches during extrusion.

Preferred particles are SiO_2 in colloidal or in chain form. These particles are very effectively incorporated by bonding into the polymer matrix. There is in principle no restriction on the diameter of the particles used. However, for achieving the object it has proven advantageous to use particles whose average diameter (generally the d_{50} value) is greater than 1 µm, preferably in the range from 1.5 to 10 µm, and particularly preferably in the range from 2 to 8 µm.

The pigmentation of the individual layers may accordingly be very different, and substantially depends on the structure of the film (layer structure) and on the requirements placed upon the film with regard to achievement of the other optical properties and production performance and processing performance.

In another advantageous embodiment, the outer layer (C) comprises, besides the polyethylene terephthalate homopolymer or polyethylene terephthalate copolymer, another polymeric component I. This component I is a polyethylene terephthalate copolymer which is composed of the condensate of the following monomers or of their derivatives capable of forming polyesters:

a) from 40 to 99 mol% of isophthalic acid;
b) from 0 to 60 mol% of at least one aliphatic dicarboxylic acid having the formula $HOOC(CH_2)_nCOOH$, where n is in the range from 1 to 11;
c) from 1 to 15 mol% of at least one sulfomonomer containing an alkali metal sulfonate group on the aromatic moiety of a dicarboxylic acid;
d) the stoichiometric amount of a copolymerizable aliphatic or cycloaliphatic glycol having from 2 to 11 carbon atoms needed to form 100 mol% of condensate;

where each of the percentages is based on the total amount of the monomers forming component I. For the detailed description of component I, see also **EP-A-0 144 878**, expressly incorporated herein by way of reference.

Component I is advantageously added in the form of a further polymeric component to the matt outer layer (C), and the proportion here may reach up to 60 % by weight. In this case, component I forms, with the other polymers present in this layer, a blend or a mixture, or else a copolymer via transesterification in the extrusion process.

For the purposes of the present invention, mixtures are mechanical mixtures which are prepared from the separate components. For this, the separate constituents are generally combined in the form of small-dimension compressed moldings, e.g. lenticular or bead-shaped pellets, and mixed with one another mechanically, using a suitable agitator. Another way of producing the mixture is to feed component I and the corresponding other polymer for the outer layer C separately to the extruder for the outer layer and to mix the materials in the extruder, or in downstream systems for conducting the melts.

For the purposes of the present invention, a blend is an alloy-like composite of the individual components which can no longer be broken down to give the original constituents. The properties of the blend are like those of a homogeneous substance, and it can therefore be appropriately characterized via suitable parameters.

The desired property combinations can in particular be achieved if the topography of the matt outer layer (C) is described by the following set of parameters:

a) the roughness of the film expressed in the form of its R_a value, is in the range from 150 to 1000 nm,

preferably from 175 to 950 nm, and particularly preferably from 200 to 900 nm. Values smaller than 150 nm have adverse effects on the degree of mattness of the surface, and values greater than 1000 nm impair the optical properties of the film.

b) the value measured for surface gas flow is in the range from 0 to 80 s, preferably in the range from 1 to 50 s. At values above 80 s, the degree of mattness of the film is adversely affected.

The base layer (B) may likewise also comprise conventional additives, such as stabilizers and/or pigments (= fillers). Examples of stabilizers advantageously used are phosphorus compounds, such as phosphoric acid or phosphoric esters.

Typical pigments (= fillers) for the base layer are the inorganic and/or organic particles given for the two outer layers, for example calcium carbonate, amorphous silica, talc, magnesium carbonate, barium carbonate, calcium sulfate, barium sulfate, lithium phosphate, calcium phosphate, magnesium phosphate, aluminum oxide, LiF, the calcium, barium, zinc, or manganese salts of the dicarboxylic acids used, carbon black, titanium dioxide, kaolin, or crosslinked polystyrene particles or crosslinked acrylate particles.

In the advantageous usage form, the film is composed of three layers, the base layer (B) and, applied to the two sides of this base layer, outer layers (A) and (C), the outer layer (A) being sealable with respect to itself and with respect to the outer layer (C).

Between the base layer (B) and the outer layers (A) and

(C) there may, where appropriate, be an intermediate layer. This may again be composed of the polymers described for the base layer or the outer layers (A) and (C). In one particularly preferred embodiment, it is composed of the polyester used for the base layer. It may also comprise the customary additives described. The thickness of the intermediate layer is generally above 0.3 µm, preferably in the range from 0.5 to 15 µm, in particular in the range from 1.0 to 10 µm and very particularly preferably in the range from 1.0 to 5 µm.

In the particularly advantageous three-layer embodiment of the film of the invention, the thickness of the outer layers (A) and (C) is generally in the range from 0.1 to 5.0 mm, preferably in the range from 0.2 to 4.5 mm, and particularly preferably in the range from 0.3 to 4.0 mm, and the thickness of the outer layers (A) and (C) here may be identical or different.

The total thickness of the film of the invention may vary within certain limits. It is from 3 to 350 µm, in particular from 4 to 300 µm, preferably from 5 to 250 µm, the layer B preferably making up from 5 to 95 % of the total thickness.

To produce the film, the polymers for the base layer (B) and the two outer layers (A) and (C) are introduced to three extruders. Any foreign bodies or contamination present may be filtered out by suitable filters from the polymer melt prior to/during extrusion. The melts are then extruded through a coextrusion die to give flat melt films, and layered one upon the other. The multilayer film is then drawn off and solidified with

the aid of a chill roll and, if desired, other rolls.

The invention also relates to a process for producing the polyester film of the invention in accordance with the coextrusion process known per se from the literature.

The procedure for this process is that the melts corresponding to the individual layers (A), (B), and (C) of the film are coextruded through a flat-film die, the resultant film is drawn off for solidification on one or more rolls, the film is then biaxially stretched (oriented), the biaxially stretched film is heat-set and, if desired, corona- or flame-treated on the surface layer intended for treatment.

The biaxial stretching (orientation) is generally carried out sequentially, and preference is given to sequential biaxial stretching in which stretching is first longitudinal (in the machine direction) and then transverse (perpendicular to the machine direction).

As is usual in coextrusion, the polymer or the polymer mixtures for the individual layers is first compressed and plasticized in an extruder, and any additives used may already be present in the polymer or the polymer mixture during this process. The melts are then simultaneously extruded through a flat-film die (slot die), and the extruded multilayer film is drawn off on one or more take-off rolls, whereupon it cools and solidifies to give a pre-film.

The biaxial orientation is generally carried out sequentially, preferably orienting first longitudinally

(i.e. in the machine direction = MD) and then transversely (i.e. perpendicularly to the machine direction = TD). This gives orientation of the molecular chains. The longitudinal orientation can be carried out with the aid of two rolls running at different speeds corresponding to the desired stretching ratio. For the transverse orientation use is generally made of an appropriate tenter frame, in which the two edges of the film are clamped and then drawn toward the two sides at an elevated temperature.

The temperature at which the orientation is carried out may vary over a relatively wide range and depends on the film properties desired. The longitudinal stretching is generally carried out at a temperature in the range from 80 to 130 °C, and the transverse stretching in the range from 90 to 150 °C. The longitudinal stretching ratio is generally in the range from 2.5:1 to 6:1, preferably from 3:1 to 5.5:1. The transverse stretching ratio is generally in the range from 3.0:1 to 5.0:1, preferably from 3.5:1 to 4.5:1. Prior to the transverse stretching, one or both surfaces of the film may be in-line coated by known processes. The in-line coating may serve, for example, to give improved adhesion of a metal layer or of any printing ink to be applied later, or else to improve antistatic performance or processing performance.

For producing a film with not only good sealing properties but also comparatively low haze, it has proven advantageous for the planar orientation Δp of the film to be less than Δp = 0.170, but particularly less than Δp = 0.168, and very particularly preferably

less than $\Delta p = 0.166$. In this case the strength of the film in the direction of its thickness is so great that when the seal seam strength is measured it is specifically the seal seam which separates, and the tear does not enter the film or propagate therein. In addition, there is then less formation, around the respective particles, of vacuols which generate haze.

The significant variables affecting the planar orientation Δp have been found to be the longitudinal and transverse stretching parameters, and also the SV value of the raw material used. The process parameters include in particular the longitudinal and transverse stretching ratios (λ_{MD} and λ_{TD}), the longitudinal and transverse stretching temperatures (T_{MD} and T_{TD}), the film web speed and the nature of the stretching, in particular that in the longitudinal direction of the machine. For example, if the planar orientation obtained with a machine is $\Delta p = 0.172$ with the following set of parameters: $\lambda_{MD} = 4.8$ and $\lambda_{TD} = 4.0$, a longitudinal stretching temperature T_{MD} of from 80 - 118 °C and a transverse stretching temperature T_{TD} of from 80 - 125 °C, then increasing the longitudinal stretching temperature T_{MD} to 80 - 125 °C or increasing the transverse stretching temperature T_{TD} to 80 - 135 °C, or lowering the longitudinal stretching ratio λ_{MD} to 4.3 or lowering the transverse stretching ratio λ_{TD} to 3.7 gives a planar orientation Δp within the desired range. The film web speed here was 340 m/min and the SV value for the material was about 730. For the longitudinal stretching, the data mentioned are based on what is known as N-TEP stretching, composed of a low-orientation stretching step (LOE, Low Orientation Elongation) and a high-orientation stretching step

(REP, Rapid Elongation Process). Other stretching systems in principle give the same ratios, but the numeric values for each process parameter may be slightly different. The temperatures given are based on the respective roll temperatures in the case of the longitudinal stretching and on infrared-measured film temperatures in the case of the transverse stretching.

In the heat-setting which follows, the film is held for from about 0.1 to 10 s at a temperature of from 150 to 250 °C. The film is then wound up in a usual manner.

After the biaxial stretching it is preferable for one or both surfaces of the film to be corona- or flame-treated by one of the known methods. The intensity of the treatment is such that it results in a surface tension in the range above 45 mN/m.

The film may also be coated in order to achieve other desired properties. Typical coatings are layers with adhesion-promoting, antistatic, slip-improving or release action. These additional layers may be applied to the film by way of in-line coating, using aqueous dispersions, prior to the transverse stretching step.

The inventive sealable film with one matt side features excellent sealability, attractive optical properties, very good handling, and very good processing performance. The sealable outer layer (A) of the film seals not only with respect to itself (fin sealing) but also with respect to the matt outer layer (C) (lap sealing).

In addition, the film has an impressive matt surface A

effective for promotional purposes, without any marked resultant impairment of (increase in) the haze of the film. Furthermore, it has been ensured that during the production of the film it is possible to reintroduce an amount in the range from 10 to 60 % by weight, based on the total weight of the film, of the chopped material (regrind) to the extrusion process, with no significant resultant adverse effect on the physical properties of the film, in particular its appearance.

The film therefore has quite excellent suitability for use in flexible packaging and specifically and particularly wherever its excellent sealing properties and its good processability can be fully utilized. A particular case here is its use in high-speed packaging machinery.

The table below (table 1) gives again the most important inventive film properties at a glance.

Table 1

	Inventive range	Preferred	Particularly preferred	Unit	Test method
OUTER LAYER A					
Minimum sealing temperature	< 120	< 115	< 110	°C	internal
Seal seam strength	> 1.0	> 1.2	> 1.4	N/15 mm	internal
Average roughness R_a	≤ 100	≤ 90	≤ 80	nm	DIN 4768, cut-off of 0.25 mm
Range of values measured for surface gas flow	20-4000	50-3500	60-3000	sec	internal
Gloss 60 °	> 100	> 110	> 120		DIN 67530
OUTER LAYER C					
Gloss 60 °	< 100	< 90	< 80		DIN 67530
Average roughness R_a	150-1000	175-950	200-900	nm	DIN 4768, cut-off of 0.25 mm

Range of values measured for surface gas flow	≤ 80	1-50	2-30	sec	internal
COF	< 0.6	< 0.45	<0.40		DIN 53375
Other film properties					
Haze	< 50	< 45	< 40	%	ASTM-D 1003-52
Planar orientation Δp	< 0.170	< 0.168	< 0.166		internal

The following test methods were utilized for the purposes of the present invention to characterize the raw material and the films:

SV value (standard viscosity)

Standard viscosity SV (DCA) is determined by a method based on DIN 53726, in dichloroacetic acid. Intrinsic viscosity (IV) is calculated as follows from standard viscosity

$$IV\ (DCA) = 6.907 \times 10^{-4}\ SV\ (DCA) + 0.063096$$

Determination of minimum sealing temperature

Hot-sealed specimens (seal seam 20 mm × 100 mm) are produced with a Brugger HSG/ET sealing apparatus, by sealing the film at different temperatures with the aid of two heated sealing jaws at a sealing pressure of 4 bar and with a sealing time of 0.5 s. From the sealed specimens test strips of 15 mm width were cut. The T-seal seam strength was measured as in the determination of seal seam strength. The minimum sealing temperature is the temperature at which a seal seam strength of at least 0.5 N/15 mm is achieved.

Seal seam strength

To determine seal seam strength, two film strips of width 15 mm were placed one on top of the other and sealed at 130 °C with a sealing time of 0.5 s and a

sealing pressure of 2 bar (apparatus: Brugger model NDS, single-side-heated sealing jaw). The seal seam strength was determined by the T-peel method.

Coefficient of friction

Coefficient of friction was determined to DIN 53 375. The coefficient of sliding friction was measured 14 days after production.

Surface tension

Surface tension was determined by what is known as the ink method (DIN 53 364).

Haze

Hölz haze was measured by a method based on ASTM-D 1003-52 but, in order to utilize the most effective measurement range, measurements were made on four pieces of film laid one on top of the other, and a 1° slit diaphragm was used instead of a 4° pinhole.

Gloss

Gloss was determined to DIN 67 530. The reflectance was measured as an optical value characteristic of a film surface. Based on the standards ASTM-D 523-78 and ISO 2813, the angle of incidence was set at 20° or 60°. A beam of light hits the flat test surface at the set angle of incidence and is reflected and/or scattered thereby. A proportional electrical variable is displayed representing light rays hitting the photoelectronic detector. The value measured is dimensionless and must be stated together with the angle of incidence.

Surface gas flow time

The principle of the test method is based on the air flow between one side of the film and a smooth silicon wafer sheet. The air flows from the surroundings into an evacuated space, and the interface between film and silicon wafer sheet acts as a flow resistance.

A round specimen of film is placed on a silicon wafer sheet in the middle of which there is a hole providing the connection to the receiver. The receiver is evacuated to a pressure below 0.1 mbar. The time in seconds taken by the air to establish a pressure rise of 56 mbar in the receiver is determined.

Test conditions:

Test area	45.1 cm^2
Weight applied	1276 g
Air temperature	23 °C
Humidity	50% relative humidity
Aggregated gas volume	1.2 cm^3
Pressure difference	56 mbar

Determination of planar orientation Δp

Planar orientation is determined by measuring the refractive indices with an Abbe refractometer in accordance with internal operations specification 24.

Preparation of specimens:

Specimen size and length:	from 60 to 100 mm
Specimen width:	corresponds to prism width of 10 mm

To determine n_{MD} and n_{α} (= n_z), the specimen to be

tested has to be cut out from the film with the running edge of the specimen running precisely in the direction TD. To determine n_{TD} and n_{α} (= n_z), the specimen to be tested has to be cut out from the film with the running edge of the specimen running precisely in the direction MD. The specimens are to be taken from the middle of the film web. Care must be taken that the temperature of the Abbe refractometer is 23 °C. Using a glass rod, a little diiodomethane (N = 1.745) or diiodomethane-bromonaphthalene mixture is applied to the lower prism, which has been cleaned thoroughly before the test. The refractive index of the mixture must be greater than 1.685. The specimen cut out in the direction TD is firstly laid on top of this, in such a way that the entire surface of the prism is covered. Using a paper wipe, the film is now firmly pressed flat onto the prism, so that it is firmly and smoothly positioned thereon. The excess liquid must be sucked away. A little of the test liquid is then dropped onto the film. The second prism is swung down and into place and pressed firmly into contact. The right-hand knurled screw is then used to turn the indicator scale until a transition from light to dark can be seen in the field of view in the range from 1.62 to 1.68. If the transition from light to dark is not sharp, the colors are brought together using the upper knurled screw in such a way that only one light and one dark zone are visible. The sharp transition line is brought to the crossing point of the two diagonal lines (in the eyepiece) using the lower knurled screw. The value now indicated on the measurement scale is read off and entered into the test record. This is the refractive index n_{MD} in the machine direction. The scale is now turned using the lower knurled screw until the range

visible in the eyepiece is from 1.49 to 1.50.

The refractive index n_α or n_z (in the direction of the thickness of the film) is then determined. To improve the visibility of the transition, which is only weakly visible, a polarization film is placed over the eyepiece. This is turned until the transition is clearly visible. The same considerations apply as in the determination of n_{MD}. If the transition from light to dark is not sharp (colored), the colors are brought together using the upper knurled screw in such a way that a sharp transition can be seen. This sharp transition line is placed on the crossing point of the two diagonal lines using the lower knurled screw, and the value indicated on the scale is read off and entered into the table.

The specimen is then turned, and the corresponding refractive indices n_{MD} and n_α ($= n_z$) of the other side are measured and entered into an appropriate table.

After determining the refractive indices in, respectively, the direction MD and the direction of the thickness of the film, the specimen strip cut out in the direction MD is placed in position and the refractive indices n_{TD} and n_α ($= n_z$) are determined accordingly. The strip is turned over, and the values for the B side are measured. The values for the A side and the B side are combined to give average refractive indices. The orientation values are then calculated from the refractive indices using the following formulae:

$\Delta n = n_{MD} - n_{TD}$

$\Delta p = (n_{MD} + n_{TD})/2 - n_z$

$n_{av} = (n_{MD} + n_{TD} + n_z)/3$

Inventive example 1

Chips composed of polyethylene terephthalate (prepared by way of the transesterification process, using Mn as transesterification catalyst, Mn concentration: 100 ppm) were dried at a temperature of 150 °C to a residual moisture level below 100 ppm, and introduced into the extruder for the base layer (B).

Chips composed of polyethylene terephthalate and of a filler were likewise introduced into the extruder for the matt outer layer (C).

Alongside this, chips were prepared from a linear polyester which is composed of an amorphous copolyester (hereinafter termed copolyester I) with 78 mol% of ethylene terephthalate and 22 mol% of ethylene isophthalate (prepared by way of the transesterification process using Mn as transesterification catalyst, Mn concentration: 100 ppm). Copolyester I was dried at a temperature of 100 °C to a residual moisture level below 200 ppm, and introduced into the extruder for the sealable outer layer (A).

A transparent, three-layer film with ABC structure and a total thickness of 12 µm was then produced via coextrusion followed by stepwise longitudinal and transverse orientation. The thickness of the individual outer layers is to be found in table 2.

Outer layer (A) was a mixture composed of:

96.0 % by weight	of copolyester I with SV 800
4.0 % by weight	of masterbatch composed of

	97.75 % by weight of copolyester I and 1.0 % by weight of ®Sylobloc 44 H (synthetic SiO_2 from Grace), and 1.25 % by weight of ®Aerosil TT 600 (fumed SiO_2 from Degussa)

Base layer (B):

100.0 % by weight	of polyethylene terephthalate with SV 800

Outer layer (C) was a mixture composed of:

40 % by weight	of polyethylene terephthalate with SV 800
60 % by weight	of masterbatch composed of 95 % by weight of polyethylene terephthalate and 5.0 % by weight of silicic acid particles (®Sylobloc 44 H from Grace) with an average particle size of 4.5 µm.

The production conditions in the individual steps of the process were:

Extrusion:	Temperatures	Layer A:	270 °C
		Layer B:	290 °C
		Layer C:	290 °C
	Die gap width:		2.5 mm
	Take-off roll Temperature:		30 °C
Longitudinal stretching:	Temperature:		80-126 °C

	Longitudinal stretching ratio:	4.0
Transverse stretching:	Temperature:	80-135 °C
	Transverse stretching ratio:	4.0
Heat-setting:	Temperature:	230 °C
Duration:		3 s

The film had the required low gloss, the low haze, and the good sealing properties. Furthermore, the film exhibits the desired handling and the desired processing performance. Tables 2 and 3 show the structure of the film and the properties achieved in films produced in this way.

Inventive example 2

In comparison with inventive example 1, the thickness of the sealable outer layer (A) was raised from 1.5 to 2.0 µm, while the film structure was otherwise identical, as was its method of production. The result was an improvement in sealing properties, and in particular the marked increase in seal seam strength.

Inventive example 3

In comparison with inventive example 1, a film of thickness 23 µm was now produced. The thickness of the sealable outer layer (A) was 2.5 µm, and that of the nonsealable outer layer (C) was 2.0 µm. The result was a further reduction in the gloss of the film and again

an improvement in the sealing properties, and in particular again a marked increase in the seal seam strength. Again, there was also a slight improvement in the handling of the film.

Inventive example 4

In comparison with inventive example 3, the copolymer for the sealable outer layer (A) was changed. Instead of the amorphous copolyester having 78 mol% of polyethylene terephthalate and 22 mol% of ethylene isophthalate, use was now made of an amorphous copolyester having 70 mol% of polyethylene terephthalate and 30 mol% of ethylene isophthalate. The polymer was processed in a twin-screw vented extruder, and did not require pre-drying. The thickness of the sealable outer layer (A) was again 2.5 µm, and that of the nonsealable layer (C) was 2.0 µm. The result was an improvement in the sealing properties, and in particular a marked increase in the seal seam strength. The pigment concentration in the sealable outer layer (A) was raised slightly in order to achieve good handling and good processing performance in the film.

Inventive example 5

In comparison with inventive example 3, the composition of the matt outer layer (C) was changed. Instead of the polyethylene terephthalate with SV 800, 20 % by weight of the polymeric component I were now added to the matt outer layer (C). The component I has the following composition:

a) 90 mol% of isophthalic acid;
b) 10 mol% of the sodium salt of sulfoisophthalic acid.

The introduction of component I into the matt outer layer further improved the haze of the film and the sealing of side (A) with respect to side (C).

Outer layer (C) was a mixture composed of:

20 % by weight	of polyethylene terephthalate with SV 800
20 % by weight	of component I
60 % by weight	of masterbatch composed of 95 % by weight of polyethylene terephthalate and 5.0 % by weight of silicic acid particles (®Sylobloc 44 H from Grace) with an average particle size of 4.5 µm.

Comparative example 1

In comparison with inventive example 1, the sealable outer layer (A) was now not pigmented. Although the result was some improvement in the sealing properties, the handling of the film and the processing performance have undergone unacceptable deterioration.

Comparative example 2

In comparison with inventive example 1, the level of pigmentation in the sealable outer layer (A) was now raised to that of the nonsealable outer layer (C). This measure improved the handling and the processing properties of the film, but there is a marked deterioration in the sealing properties.

Comparative example 3

In comparison with inventive example 1, the level of pigmentation in the nonsealable outer layer (C) was now markedly reduced. The result was marked impairment of

the handling of the film and of its processing performance.

Comparative example 4

Example 1 from EP-A-0 035 835 was repeated. The sealing performance of the film, its handling, and its processing performance are poorer than in the inventive examples.

Table 2

Example	Film thickness µm	Film structure	Layer thicknesses µm			Pigments in layers			Average pigment diameter in layers µm			Pigment concentrations ppm		
			A	B	C	A	B	C	A	B	C	A	B	C
IE 1	12	ABC	1.5	9	1.5	Sylobloc 44 H Aerosil TT 600	none	Sylobloc 44	2.5 0.04		4.5	300 375	0	30 000
IE 2	12	ABC	2.0	8.5	1.5	Sylobloc 44 H Aerosil TT 600	none	Sylobloc 44	2.5 0.04		4.5	300 375	0	30 000
IE 3	23	ABC	2.5	18.5	2.0	Sylobloc 44 H Aerosil TT 600	none	Sylobloc 44	2.5 0.04		4.5	300 375	0	30 000
IE 4	23	ABC	2.5	18.5	2.0	Sylobloc 44 H Aerosil TT 600	none	Sylobloc 44	2.5 0.04		4.5	400 500	0	30 000
IE 5	23	ABC	2.5	18.5	2.0	Sylobloc 44 H Aerosil TT 600	none	Sylobloc 44	2.5 0.04		4.5	400 500	0	30 000
CE 1	12	ABC	1.5	9	1.5	none	none	Sylobloc 44 H Aerosil TT 600			2.5 0.04		0	1200 1500
CE 2	12	ABC	1.5	9	1.5	Sylobloc 44 H Aerosil TT 600	none	Sylobloc 44 H Aerosil TT 600	2.5 0.04		1200 1500	300 375	0	1200 1500
CE 3	12	ABC	1.5	9	1.5	Sylobloc 44 H Aerosil TT 600	none	Sylobloc 44 H Aerosil TT 600	2.5 0.04		2.5 0.04	300 375	0	600 750
CE 4	15	AB	2.25	12.75		Gasil 35	none		3			2500	0	

Table 3

Ex.	Minimum sealing temperature °C	Seal seam strength N/15 mm	Seal seam strength N/15 mm	Coefficient of friction COF	Average roughness R_a nm		Values measured for gas flows		Δp	Gloss		Haze	Winding performance and handling	Processing performance
	Side A with respect to side A	Side A with respect to side A	Side A with respect to side C	Side C with respect to side C	Side A	Side C	Side A	Side C		Side A	Side C			
IE 1	100	2.0	1.3	0.40	25	230	1200	10	0.165	140	55	45	++	++
IE 2	98	2.7	1.5	0.40	26	230	1280	10	0.165	140	55	45	++	++
IE 3	95	3.0	1.8	0.40	23	230	1110	10	0.165	130	50	47	++	++
IE 4	85	3.3	2.0	0.40	23	230	1300	10	0.165	130	50	47	++	++
IE 5	85	3.3	2.0	0.40	23	250	1300	8	0.165	130	45	35	++	++
CE 1	98	2.1		0.45	10	65	10 000	80	0.165	160	170	1.5	-	-
CE 2	110	1.0		0.45	65	65	80	80	0.165	130	170	2.8	-	-
CE 3	100	2.0		0.45	25	37	1200	150	0.165	160	190	1.5	-	-
CE 4	115	0.97		> 2	70	20	50	> 5000				12	-	-

Key to winding performance, handling, and processing performance of films:

++: no tendency to adhere to rolls or other mechanical parts, no blocking problems on winding or during processing on packaging machinery, low production costs

+: moderate production costs

-: tendency to adhere to rolls or other mechanical parts, blocking problems on winding and during processing on packaging machinery, high production costs due to complicated handling of film in machinery